



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US83/01634 <b>(22) International Filing Date:</b> 19 October 1983 (19.10.83)  <b>(31) Priority Application Numbers:</b> 440,764 518,583  <b>(32) Priority Dates:</b> 10 November 1982 (10.11.82) 29 July 1983 (29.07.83)  <b>(33) Priority Country:</b> US  <b>(71) Applicant:</b> INMONT CORPORATION [US/US]; 1255 Broad Street, Clifton, NJ 07015 (US).  <b>(72) Inventor:</b> PANUSH, Sol ; 28670 Oak Crest Drive, Far- mington Hills, MI 48018 (US).  <b>(74) Agent:</b> GWINNELL, Harry, J.; Patent Department, United Technologies Corporation, Hartford, CT 06101 (US).		<b>(81) Designated States:</b> AU, BE (European patent), BR, DE (European patent), FR (European patent), GB (Euro- pean patent), JP.  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> PEARLESCENT AUTOMOTIVE PAINT COMPOSITIONS  <b>(57) Abstract</b>  A coating composition comprising a thermoplastic or thermosetting resin material containing a pigment composi- tion. The pigment composition contains about 5 to about 90% iron oxide encapsulated mica particles. When used in a mul- ti-coat coating process as a basecoat overcoated with a transparent overcoat, an article is produced which not only pro- duces improved pearlescent color effects, but is stable to the elements as well.		

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- 1 -

## Description

## Pearlescent Automotive Paint Compositions

## Technical Field

5 The field of art to which this invention  
pertains is coating compositions, coating methods,  
and the resultant coated articles.

## Background Art

10 Multicoat coating systems are now well  
known in the coating industry. U. S. Patent 3,639,147  
describes such a system for use as an automotive  
paint. When multicoat coating systems have  
previously been applied, it has been difficult  
to obtain coatings having uniformly high gloss  
while at the same time maintaining good color,  
15 and pearlescent features.

In the automotive industry two significant  
systems are utilized in the painting of automobiles.  
One topcoat system which utilizes thermoplastic  
resins is known as the acrylic lacquer system.  
20 In this system the base polymers are the homopolymers  
of methyl methacrylate and copolymers of methyl  
methacrylate and acrylic acid, methacrylic acid,  
alkyl esters of acrylic acid or methacrylic  
acid, vinyl acetate, acrylonitrile, styrene and  
25 the like. The acrylic lacquer topcoats have been  
acknowledged to have outstanding aesthetic  
properties. Another outstanding topcoat system  
used in the automotive industry is the thermosetting  
acrylic resins as described in U. S. Patent 3,375,227,  
30 issued March 26, 1968.



- 2 -

These topcoat systems have outstanding chemical resistance, outstanding resistance to cracking and crazing among other outstanding properties, but to the expert paint formulator, the thermosetting acrylic resins have not, in the past, quite provided the aesthetic properties obtained in the acrylic lacquer systems. In these systems a pigmented base coat composition is applied to the metal substrate to hide metallic blemishes and provide the aesthetically pleasing colors desired followed by the application of an unpigmented layer of polymer which imparts a "deep" color appearance to the base coat and durability to this pigmented base coat. This system, however, is not without its problems. Aesthetic quality of the coating is totally dependent on the application of the base coat. The clear top coat magnifies any weakness in this base coat including the highlighting of any color deficiencies of the base coat. The clear coat also acts as a magnifying mirror for ultraviolet radiation which can accelerate rather than retard any degradation of the base coat due to exposure to ultraviolet radiation. In addition, many of these coating systems in use today utilize metal particles in the base coat to provide an aesthetically pleasing metallic appearance. However, problems have occurred with



- 3 -

the use of metallic pigments resulting in color loss in the base coat.

5 In order to overcome the deficiencies of the metallic pigments, pearlescent pigments have been considered for use in the basecoat, either with or in place of conventional pigments. However, use of these pigments have included such problems as moisture sensitivity, problems with large particle sizes, application control problems, 10 problems with color travel (face to flop), sensitivity to sunlight, subtractive color effects, and quality control problems. Both with metallic pigments and with the many pearlescent pigments available problems such as ghosting, modeling, 15 silking, alligatoring, telegraphing, etc. have also been observed. Use of a three coat system can correct some of these problems. However, this is not an acceptable solution in the industry. Additional coating steps can only aggravate already 20 insurmountable application control problems.

Accordingly, what is needed in this art are improved coating compositions and coating systems which are aesthetically pleasing as well as durable and easy to control.

## 25 Disclosure of Invention

A coating composition is disclosed comprising a thermoplastic or thermosetting polymer composition containing about 1% to about 20% by weight of a pigment composition. The pigment composition 30 contains about 5% to about 90% by weight of iron oxide encapsulated mica particles. The mica particles are about 5 to about 60 microns nominal longitudinal dimension and have a thickness of about 0.25 to about 1 micron. The encapsulating 35 layer constitutents about 10% to about 85% by weight of the particle weight. The iron



- 4 -

oxide layer can contain other constituents such as minor amounts of chromium hydroxide and titanium dioxide.

Another aspect of the invention includes  
5 a substrate material having coated thereon a multilayer coating system comprising at least one basecoat and at least one topcoat. The basecoat is a pigmented resin containing iron oxide encapsulated mica particles as described  
10 above. The topcoat is a transparent thermoplastic or thermosetting material.

Another aspect of the invention includes a method of coating a substrate by depositing a layer of the pigmented, iron oxide encapsulated mica  
15 containing polymer composition described above and applying a layer of transparent thermoplastic or thermosetting resin thereon.

By utilizing the compositions and processes of this invention, not only are truer, more  
20 brilliant colors produced but colors durable to the elements are produced as well.

The foregoing, and other features and advantages of the present invention, will become more apparent from the following description.  
25

#### Best Mode for Carrying Out the Invention

While any substrate material can be coated with the coating compositions according to the present invention, including such things as  
30 glass, ceramics, asbestos, wood, and even plastic material depending on the specific drying and/or curing requirements of the particular composition, the coating system of the present invention is particularly adapted for metal substrates,  
35 and specifically as an automotive paint finish



system. The substrate may also be bare substrate material or can be conventionally primed, for example to impart corrosion resistance. Exemplary metal substrates include such things as steel, aluminum, copper, magnesium, alloys thereof, etc. The components of the composition can be varied to suit the temperature tolerance of the substrate material. For example, the components can be so constituted for air drying (i.e. ambient), low temperature cure (e.g. 150°F-180°F), or high temperature cure (e.g. over 180°F).

The basecoat material, i.e. the pigmented polymer layer closest to the substrate, comprises any suitable film forming material conventionally used in this art including acrylics, alkyds, polyurethanes, polyesters and aminoplast resins. Although the basecoat can be deposited out of an aqueous carrier, it is preferred to use conventional volatile organic solvents such as aliphatic, cycloaliphatic and aromatic hydrocarbons, esters, ethers, ketones and alcohols including such things as toluene, xylene, butyl acetate, acetone, methyl isobutyl ketone, butyl alcohol, etc. When using volatile organic solvents, although it is not required, it is preferred to include from about 2% to about 50% by weight of a cellulose ester and/or wax (e.g. polyethylene) which facilitates quick release of the volatile organic solvent resulting in improved flow or leveling out of the coating. The cellulose esters used must be compatible with the particular resin systems selected and include such things as cellulose nitrate, cellulose propionate, cellulose butyrate, cellulose acetate butyrate, cellulose acetate propionate, and mixtures thereof. The cellulose esters when used are preferably used in about 5% to about 20% by weight based on film forming solids.



The acrylic resins in the basecoat may be either thermoplastic (acrylic lacquer systems) or thermosetting. Acrylic lacquers such as are described in U. S. Patent 2,860,110 are one type of film forming composition useful according to this invention in the basecoat. The acrylic lacquer compositions typically include homopolymers of methyl methacrylate and copolymers of methyl methacrylate which contain among others, acrylic acid, methacrylic acid, alkyl esters of acrylic acid, alkyl esters of methacrylic acid, vinyl acetate, acrylonitrile, styrene and the like.

When the relative viscosity of the acrylic lacquer polymer is less than about 1.05, the resulting films have poor solvent resistance, durability and mechanical properties. On the other hand, when the relative viscosity is increased above the 1.40 level, paints made from these resins are difficult to spray and have high coalescing temperatures.

Another type of film forming material useful in forming the basecoat of this invention is a combination of a crosslinking agent and a carboxy-hydroxy acrylic copolymer. Monomers that can be copolymerized in the carboxy-hydroxy acrylic co-polymer include esters of acrylic and methacrylic acid with alkanols containing 1 to 12 carbon atoms, such as ethyl acrylate, methyl methacrylate, butyl acrylate, butyl methacrylate, 2-ethylhexyl acrylate, lauryl methacrylate, benzyl acrylate,





- 7 -

cyclohexyl methacrylate, and the like. Additional monomers are acrylonitrile, methacrylonitrile, styrene, vinyl toluene, alpha-methyl styrene, vinyl acetate, and so forth. These monomers  
5 contain one polymerizable ethylenically unsaturated group and are devoid of hydroxyl and carboxylic groups.

The crosslinking agents used in combination with the hydroxy-carboxy copolymers are those  
10 compositions which are reactive with hydroxy and/or carboxylic acid groups. Examples of such crosslinking agents are polyisocyanates (typically di- and/or tri- isocyanates) polyisocyanates, polyepoxides and aminoplast resins. Particularly  
15 preferred crosslinking agents are the aminoplast resins.

The polyisocyanates when reacted with hydroxyl bearing polyester or polyether or acrylic polymers will yield urethane films useful in the process  
20 of this invention in both the basecoat and topcoat. The isocyanate (-NCO) - hydroxyl (-OH) reaction takes place readily at room temperature, so that ambient and low temperature cure is possible.

Among other basecoats which are typically used  
25 in the processes of the present invention are those commonly known as alkyd resins which are defined to include fatty acid or oil containing esterification products. The methods for preparing these resins are well known in the art.

30 The preferred alkyd resins useful in this invention are those containing from about 5 to about 65 weight percent of a fatty acid or oil and having an hydroxyl equivalent to carboxy equivalent ratio of from about 1.05 to 1.75. Alkyd resins having



less than about 5% fatty compound are classified as the "oil-less" alkyd resins or polyester resins described hereinafter. On the other hand, alkyd resins containing greater than 65% of a fatty compound exhibit poor baking properties, poor chemical resistance and unsatisfactory adhesion to either the basecoat or the substrate. When the hydroxyl to carboxyl equivalent ratio is less than about 1.05 gelation can result during polymer preparation while resins prepared having a ratio in excess of 1.75 have low molecular weights and therefore poor chemical resistance.

These alkyd resins can also be used as the topcoat of this invention. When this is the case it is preferred that the oil or fatty acid portion of the alkyd resin contain a light colored baking oil or fatty acid such as coconut or dehydrated castor oils or fatty acids. Furthermore, when these resins are used as topcoats they can be reacted with various acrylic or ethylenically unsaturated monomers as described above to produce vinyl modified alkyd resins.

Curing of these alkyd resins can be accomplished by blending with any of the previously described crosslinking agents in the same weight ratios as are used with carboxy-hydroxy copolymers.

Included among the various fatty acids and oils useful in preparing these alkyd resins are the fatty acids derived from the following oils; castor, dehydrated castor, coconut, corn, cottonseed, linseed, oticica, perilla, poppyseed, safflower, soybean, tung oil, etc., and the various rosins containing tall oil fatty acids. Useful polyols



- 9 -

include the various glycols, such as ethylene glycol, propylene glycol, neopentyl glycol, butylene glycol, 1, 4 butanediol, hexylene glycol, 1,6 hexanediol, the polyglycols such as diethylene glycol or triethylene glycol, etc.; the triols such as glycerine, trimethylol ethane, trimethylol propane, etc., and other higher functional alcohols such as pentaerythritol, sorbitol, mannitol, and the like. Acids useful in preparing the alkyd resins of this invention include mono-functional acids such as rosin acids, benzoic acid, para tertiary butyl benzoic acid and the like; the polyfunctional acids such as adipic acid, azelaic acid, sebacic acid, phthalic acid or anhydride, isophthalic acid, terephthalic acid, dimerized and polymerized fatty acids, trimellitic acid, and the like.

Yet another useful basecoat is prepared using non-aqueous dispersions such as are described in U. S. Patents 3,050,412; 3,198,759; 3,232,903; 3,255,135. Typically these dispersions are prepared by polymerizing a monomer such as methyl methacrylate in the presence of a solvent in which polymers derived from the above monomer are insoluble and a precursor which is soluble in the solvent. Non-aqueous dispersions can have a relative solution viscosity as previously defined of about 1.05 to 3.0. Dispersions having a relative solution viscosity in excess of about 3.0 are difficult to spray and have high coalescence temperatures while dispersions with a relative solution viscosity less than about 1.05 have poor resistance, durability



and mechanical properties. The monomers useful in preparing the above dispersed copolymers or homopolymers are those listed previously as useful in forming the carboxy-hydroxy acrylic  
5 copolymers.

In another instance the basecoat film can be produced from resins known as polyesters or "oil-less" alkyd resins. These resins are prepared by condensing non-fatty containing polyols and  
10 polyacids. Included among the useful polyacids are isophthalic acid, phthalic acid or anhydride, terephthalic acid, maleic acid or anhydride, fumaric acid, oxalic acid, sebacic acid, azelaic acid, adipic acid, etc. Mono basic acids such as  
15 benzoic, para tertiary butyl benzoic and the like can also be utilized. Among the polyalcohols are the diols or glycols such as propylene glycol, ethylene glycol, butylene glycol, 1, 4 butanediol, neopentyl glycol, hexalene glycol, 1,6-hexanediol,  
20 and the like; the triols such as trimethylol ethane, trimethylol propane and glycerine and various other higher functional alcohols such as pentaerythritol.

Any of the above-recited polymers may be used as the topcoat, as long as it provides a  
25 transparent film. The term "transparent film" is defined as a film through which the basecoat can be seen. It is preferred that the transparent film contain a UV absorbing compound and/or a hindered amine UV stabilizer and be substantially  
30 colorless so that the full polychromatic and aesthetic effect of the basecoat is not substantially decreased. However, in some instances, desirable and unique styling effects can be obtained by the addition of contrasting or complementary colors to



the topcoat. The outstanding feature of the topcoat is the significant improvement in the durability which is provided to the overall coating composition. The use of the topcoat which covers the pigments in the basecoat also provides the opportunity to utilize the iron oxide mica encapsulated mica pigments of the present invention in the topcoat.

Utilizing the compositions of the present invention offers a means of combining the desirable properties of a combination of resin systems. For example, in automotive finishes the pigment control properties of acrylic lacquers can be combined with the chemical resistance properties of thermosetting acrylic resins by applying a thermosetting acrylic clear coat over a pigmented thermoplastic acrylic lacquer base coat (although acrylic lacquers may be used for both layers). Likewise, in appliance finishes the chemical resistance of polyester resins can be combined with the lower cost of thermosetting acrylic resins by applying a polyester clear topcoat over a pigmented thermosetting acrylic base coat. Although any of the above-mentioned thermoplastic materials may be used to form the transparent topcoat, better durability is achieved if the topcoat is one of the above-cited thermosetting materials, i.e. the material containing the cross-linking agents.

In all instances where the above methods and compositions are used extremely high gloss films result. In fact, where with normal two coat system a 60° gloss in excess of 90-95 is difficult to obtain, using the process of this invention gloss readings in excess of 100 are readily obtained.



The iron oxide encapsulated mica pigments according to the present invention are commercially available from the Mearl Corporation and EM Chemicals, and range in color from golden bronze at the thinnest iron oxide encapsulation through copper, to red at the thickest iron oxide encapsulation. The iron oxide coatings on these pigments being transparent act as natural ultra-violet absorbers. For additional exterior durability (e.g. exposure to the sun) minor amounts of other additives such as chromium hydroxide and titanium dioxide may be included in the iron oxide encapsulation layer. It should also be noted that other high temperature stable metal oxides (such as copper, calcium, cadmium, cobalt, barium, strontium, manganese, magnesium and lithium) can be substituted in whole or in part for the encapsulating iron oxide. The iron oxide encapsulation layer is generally in the molecular range of thicknesses representing about 10% to about 85% by weight of the total weight of the encapsulated mica particle, preferably about 20% to about 60%, and typically about 29% to about 48% by weight. If additives such as titanium dioxide or chromium hydroxide are used as part of the encapsulation layer they are generally present in an amount of about 1% to about 35% by weight, and typically about 2% to about 5% for the titanium dioxide, and about 0.1% to about 3.5% by weight for the chromium hydroxide, based on total weight of the encapsulated particle.



- 13 -

The uniformity of shape (platelet) and smoothness of the iron oxide encapsulated mica pigment according to the present invention (e.g. as compared to the highly fragile, three dimensional and complicated configuration of aluminum flake, a standard in the automotive paint industry) eliminates the problem of color drift due to the shear forces (yielding fragmentation problems) in the handling (overhead pumping facilities) and application problems of ghosting, mottling, silkiness and repair color matching.

The iron oxide encapsulated mica pigments are optically pearlescent because their transparency permits light to pass through the particle resulting in a multiplicity of refractions and reflections. In addition to their pearlescence these pigments have inherent hiding capabilities, are additive colors and retain a metallic appearance (face to flop color travel). These features provide the capability of producing colors with depth, clarity and chromaticity not previously attainable with conventional metals (i.e. aluminum) and previous pearlescent pigments (natural or synthetic) which produced colors lacking depth and chromaticity or color travel due to the aesthetics of the metal component (aluminum) or pearl. These colorless and opaque pigments reduced the value (color intensity) of the "true" colored pigments be they organic or inorganic resulting in gray-cloudy-low chroma colors. The addition of the iron oxide encapsulated mica pigments provides hiding and depth to travel with the aesthetically pearlescent appearance.



- 14 -

The iron oxide encapsulated mica pigments are carefully screened and controlled particles, all within about 5 microns to about 60 microns (preferably about 5 microns to about 45 microns, and typically about 5 microns to about 35 microns) in their largest dimension, and about 0.25 micron to about 1.0 micron in thickness. The closely controlled particle size provides the transparent, translucent, reflective and refractive features establishing improved aesthetic and physical properties of these coatings through careful selection and blending of these pigments. In admixture with conventional colored pigments (organic or inorganic) or dyes (natural or synthetic) unique pearlescent colors can be produced. Blending complimentary colors (e.g. red iron oxide encapsulated mica with red pigments or dyes) will produce deep chromatic colors while blending contrasting colors (e.g. bronze iron oxide encapsulated mica with red or blue pigments or dyes) will produce highly chromatic colors with unique undertones. Regardless of the selection, the final (clear coated) enamel will have improved color durability, greater moisture resistance and greater acid resistance than coatings prepared with conventional metals or synthetic pearls.

The amount of pigment in the basecoat generally comprises about 1% to about 20% by weight, preferably about 7.5% to about 15% and typically about 10% by weight. About 5% to about 90% of this pigment is the iron oxide encapsulated mica according to the present invention, preferably about 25% to about 70% and typically about 60% by weight.





- 15 -

Both the basecoat and the topcoat can be applied by any conventional method in this art such as brushing, spraying, dipping, flow coating, etc. Typically spray application is used, especially for automotive finishing. Various types of spraying can be utilized such as compressed air spraying electrostatic spraying, hot spraying techniques, airless spraying techniques etc. These can also be done by hand or by machine.

As stated above prior to application of the coating materials of the present invention a conventional corrosion resistant primer has already been applied. To this primed substrate is applied the basecoat. Prior to application of the coating materials of the present invention a conventional corrosion resistant primer typically has already been applied. To this primed substrate is applied the basecoat. The basecoat is typically applied from about 0.4 mil to about 2.0 mil and preferably about 0.5 mil to about 0.8 mils. This thickness can be applied in a single coating pass or a plurality of passes with very brief drying ("flash") between applications of coats.

Once the basecoat has been applied the transparent overcoat is applied after allowing the basecoat to flash at ambient temperature for about 30 seconds to about 10 minutes, preferably about 1 to about 3 minutes. While the basecoat can be dried for longer periods of time, even at higher temperatures, a much improved product is produced by application of the transparent topcoat after only a brief flash. Some drying out



- 16 -

of the basecoat is necessary to prevent total mixing of the basecoat and topcoat. However, a minimal degree of basecoat-topcoat interaction is desirable for improved bonding of the coatings.

5 The topcoat is applied thicker than the basecoat (preferably about 1.8 to 2.3 mils) and can also be applied in a single or multiple pass. It should also be noted that the iron oxide encapsulated mica pigments are applicable at low levels in the

10 clear coat and in basecoats with tinted clear topcoats. Both applications produce unique aesthetics while retaining the pearlescent appearance.

Pigment control is retained in the basecoat

15 while it is being overcoated. This is evidenced by lack of "strike-in" or migration of the two films (the basecoat and topcoat) into each other. When "strike-in" occurs, pigments move from the basecoat into the topcoat, the film compositions

20 become intermixed at the interface and the baked coating composition has a dusty appearance rather than a clear "depth" appearance. By this invention substantially no "strike-in" occurs, and the coatings have outstanding clarity and depth. However,

25 sufficient wetting takes place at the interface so that no problems of delamination and solvent release from either coating are obtained.

Once the topcoat is applied the system is again flashed for 30 seconds to 10 minutes and

30 the total coatings are then baked at temperature sufficient to drive off all of the solvent in the case of thermoplastic layers and at temperatures sufficient to cure and cross-link in the case of thermosetting layers. These temperatures can

35 range anywhere from ambient temperature to about



- 17 -

400°F. Typically in the case of thermosetting material temperatures of about 225°F to about 280°F (e.g. 250°F) are used, (e.g. for about 30 minutes).

5       The following examples are illustrative of the principles and practice of this invention although not limited thereto. Parts and percentages where used are parts and percentages by weight.

10       Three iron oxide encapsulated mica pigments are used in the following examples. The Golden Bronze Richelyn <sup>TM</sup> (Inmont) pigment comprises 62% - 68% mica, 29% - 35% iron oxide ( $\text{Fe}_2\text{O}_3$ ), 3% - 5% titanium dioxide ( $\text{TiO}_2$ ) and 0.3% - 0.9% chromium hydroxide ( $\text{Cr}(\text{OH})_3$ ). The Red Richelyn pigment comprises 49% - 55% mica, 42% - 48% iron oxide, 2% - 4% titanium dioxide, and 0.3% to 0.9% chromium hydroxide. The Copper Richelyn pigment comprises 51% - 57% mica, 20   40% - 46% iron oxide, 3% - 5% titanium dioxide and 0.3% - 0.9% chromium hydroxide. All percents are by weight based on the total weight of the encapsulated pigment. The Golden Bronze Richely pigment contains a relatively thin layer of iron 25   oxide encapsulation, the Red Richelyn pigment contains a relatively thick layer of iron oxide encapsulation, and the Copper Richelyn pigment contains a layer of iron oxide encapsulations which is somewhere in between.



- 18 -

Example 1

A medium brown basecoat polymer composition was prepared by blending 144 parts of the copolymer formed by reacting 47 parts of butyl methacrylate, 37 parts of styrene, 15.75 parts of hydroxypropyl methacrylate and 0.25 part of methacrylic acid with 176 parts of xylene and butanol (in a weight ratio of 85/15). A pigment base was prepared by blending 36.92 parts of Exterior Golden Bronze Richlyn pigment with 24.61 parts aluminum and 38.45 parts Transparent Yellow Oxide and 0.02 part Carbon Black. This pigment base was blended with the basecoat polymer composition in an amount representing 7.5% by weight of the composition to form the basecoat paint composition.

Bonderized steel panels primed with a cured corrosion resistant primer were sprayed with the basecoat paint composition to a film thickness of 0.5 mil on dry film basis. After a flash of approximately 2 minutes at room temperature, an additional 0.5 mil film of the basecoat paint composition again as measured on a dry film basis was applied by spraying. After a 2 minute flash at room temperature, a transparent top coating was applied by spraying to a film thickness on dry basis of 1 mil. The transparent topcoating composition was prepared by blending 144 parts of the copolymer solution described above at 45 percent nonvolatiles with 58 parts of 60 percent nonvolatile solution of butylated methylol melamine. This topcoating composition was applied by spraying to a dry film thickness of 2 mils over the basecoat described in Example 1 applied to a metal substrate. After a 2-5 minute flash time at room temperature, the coating was baked at 190°F for 15 minutes.



- 19 -

The coating was then sanded with 600 sand paper and water to smooth the coating and to remove surface imperfections. After rinsing with mineral spirits, the coating was baked at 250°F for 30 minutes. The resulting coating had reflowed before curing, the sanding marks had disappeared and the coating also had a pleasing aesthetic appearance, exhibiting superior polychromatic effects and aluminum control.

The following pigment combinations were used in the composition of the basecoat according to Example 1. In all cases the resultant coating had excellent properties for use as an automotive paint.

The first number (next to the percent pigment) represents the total pigment content of the respective paint composition.

Example 2French Vanilla

20	% Pigment	7.478
	58.81	Golden Bronze Richelyn
	33.90	Aluminum
	4.65	Transparent Yellow Oxide
	2.64	Transparent Red Oxide
25	100.00	

Example 3Medium Brown

	% Pigment	5.677
	85.86	Golden Bronze Richelyn
30	2.27	Violet RL Carbazole Violet
	11.87	Monarch 900 Carbon Black
	100.00	

Example 4Dark Plum

35	% Pigment	3.1
	48.39	Rouge Flambe Red Richelyn
	51.61	Orasol RL (Black Orasol)
	100.00	



Example 5Turquoise

	% Pigment	5.688	
	54.94	Golden Bronze Richelyn	
5	40.65	Phthalocyanine Blue	
	4.41	Monarch 900 Carbon Black	
	<u>100.00</u>		

Example 6Rich Gold

10	% Pigment	7.117	
	79.04	Golden Bronze Richelyn	
	8.78	Aluminum	
	<u>12.18</u>	Gold Monastral	
	<u>100.00</u>		

Example 7Dark Russet

	% Pigment	6.671	
	51.52	Golden Bronze Richelyn	
	46.59	Perylene	
20	<u>1.89</u>	Monarch 900 Carbon Black	
	<u>100.00</u>		

Example 8Golden Green

	% Pigment	3.723	
	61.46	Golden Bronze Richelyn	
25	5.72	Aluminum	
	19.31	Phthalocyanine Green	
	<u>13.51</u>	Monarch 900 Carbon Black	
	<u>100.00</u>		

Example 9Golden Brown

	% Pigment	5.48	
	57.2	Copper Richelyn	
	31.4	HFR Brown	
35	<u>11.4</u>	Monarch 900 Carbon Black	
	<u>100.00</u>		



- 21 -

Example 10Copper Brown

	% Pigment	5.48	
	45.6	Copper Richelyn	
5	45.3	HFR Brown	
	9.1	Monarch 900 Carbon Black	
	<u>100.00</u>		

Example 11Dark Gray

10	% Pigment	4.86	
	51.65	Red Richelyn	
	10.29	Monarch 900 Carbon Black	
	38.06	Phthalocyanine Green	
	<u>100.00</u>		

15

Example 12Medium Plum

	% Pigment	6.13	
	56.12	Red Richelyn	
	39.80	RR Indanthrene Blue	
20	4.08	Monarch 900 Carbon Black	
	<u>100.00</u>		

Example 13Medium Blue

	% Pigment	5.69	
25	55.01	Red Richelyn	
	4.39	Monarch 900 Carbon Black	
	40.60	Phthalocyanine Blue	
	<u>100.00</u>		

Example 1430 Candy Apple Red

	% Pigment	6.68	
	51.50	Red Richelyn	
	1.95	Monarch 900 Carbon Black	
	46.55	Perylene	
35	<u>100.00</u>		



- 22 -

Example 15Mustard

	% Pigment	7.5	
5	56.93	Red Richelyn	
	43.07	Yellow FR (Flavanthrone	
	<u>100.00</u>	Yellow)	

Example 16Copper Maroon

	% Pigment	4.37	
10	57.21	Red Richelyn	
	31.35	Monastral Maroon	
	11.44	Monarch 900 Carbon Black	
	<u>100.00</u>		

Example 1715 Medium Russet

	% Pigment	7.18	
	47.91	Red Richelyn	
	8.77	Aluminum	
20	43.32	Perrindo Maroon	
	<u>100.00</u>		

Example 18Cherry Red

	% Pigment	5.5	
25	45.45	Red Richelyn	
	9.10	Monarch 900 Carbon Black	
	45.45	Quinacridone Red	
	<u>100.00</u>		





The compositions and processes according to the present invention provide many improvements over the paint compositions and processes of the prior art. Pearlescent color effects at least  
5 equivalent to the use of metallic particles are produced without the need for metal particles and the application and stability problems associated with them. Novel color effects can be produced. Better hiding of surface defects can be produced.  
10 Color, hiding, fineness of particle size and reflectance not available with other pearlescent pigments are produced while maintaining the appealing and desirable soft, lustrous appearance characteristic of pearlescent. Blending with  
15 organic and/or inorganic pigments (including metal particles) is possible with enhancement of aesthetic effects produced. Weather durable color effects are produced.

The applied compositions are not moisture sensitive,  
20 use relatively small particle size, are less sensitive to criticality of applications, maintain color trueness at all angles (face to flop color travel), can withstand the elements (i.e. sun exposure), do not operate with subtractive color  
25 effects when mixed with other pigments, allow low bake repair color matching, and resist settling and chemical (e.g. acid rain) attack.

It should be noted that while the compositions of the present invention are particularly adapted  
30 for original equipment manufacture coatings for automobiles, one of their advantages is the low bake matching use as refinish compositions as



- 24 -

well. Whereas in original equipment manufacture the disclosed cellulose esters are typically used, such are not universally required for example in refinish compositions. Also, where  
5 the thermosetting polymer embodiments are preferred in the original equipment manufacture, in refinish either low temperature cure thermosetting materials (e.g. 150° to 180°F) or ambient temperature cure thermosetting or thermoplastic materials are  
10 preferred.

Although this invention has been shown and described with respect to detailed embodiments thereof, it will be understood by those skilled in the art that various changes in form and detail  
15 thereof may be made without departing from the spirit and scope of the claimed invention.



- 25 -

## Claims

1. A pearlescent coating composition comprising a thermoplastic or thermosetting resin containing about 1% to about 20% by weight of a pigment composition, wherein the pigment composition comprises about 5% to about 90% by weight of iron oxide encapsulated mica particles, the mica particles being about 5 microns to about 60 microns nominal longitudinal dimension and having a thickness of about 0.25 micron to about 1 micron, the iron oxide encapsulation representing about 10% to about 85% by weight of the total weight of the particle.

2. The composition of claim 1 wherein the iron oxide encapsulation additionally contains about 1% to about 35% titanium dioxide and about 0.1% to about 3.5% chromium hydroxide, based on total weight of the particle.

3. A substrate material coated with at least two layers of polymer including a basecoat comprising a pigmented thermosetting or thermoplastic resin and a transparent topcoat on the basecoat, wherein the pigment in the basecoat comprises iron oxide encapsulated mica particles, the mica particles being about 5 microns to about 60 microns nominal longitudinal dimension and having a thickness of about 0.25 micron to about 1 micron, the oxide encapsulation representing about 10% to about 85% by weight of the total weight of the particle.



4. The article of claim 3 wherein the substrate is metal, and the oxide encapsulation additionally contains about 1% to about 35% by weight titanium dioxide and about 0.1% to about 3.5% chromium hydroxide based on total weight of the particle.

5. A method of coating a substrate with multiple layers of polymer comprising applying at least one layer of a basecoat of pigmented thermosetting or thermoplastic resin to the substrate, applying at least one layer of a transparent thermosetting or thermoplastic topcoat on the basecoat, and drying or curing the applied coatings, the pigment comprising iron oxide encapsulated mica particles, the mica particles being about 5 microns to about 60 microns nominal longitudinal dimension and having a thickness of about 0.25 micron to about 1 micron, the iron oxide encapsulation representing about 10% to about 85% by weight of the total weight of the particle.

6. The method of claim 5 wherein the substrate is metal and the iron oxide encapsulation additionally contains about 1% to about 35% by weight titanium dioxide and about 0.3% to about 0.9% chromium hydroxide based on the total weight of the particle.



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US83/01634

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (If several classification symbols apply, indicate all) <sup>3</sup>		
According to International Patent Classification (IPC) or to both National Classification and IPC		
INT. CL. B05D 3/02, 5/06, 7/14, 7/16		
U.S. CL. 523/171, 260/998.17, 427/402, 409, 388.1, 388.2,		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>4</sup>		
Classification System	Classification Symbols	
U.S.	523/171, 260/998.17, 427/402, 409, 388.1, 388.2, 388.3, 388.4, 524/31, 32, 33, 37, 38, 39, 40, 431, 106/288R, 288B, 291, 304, DIG 3	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>5</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT</b> <sup>14</sup>		
Category *	Citation of Document, <sup>16</sup> with indication, where appropriate, of the relevant passages <sup>17</sup>	Relevant to Claim No. <sup>18</sup>
A	US, A, 3,904,421, PUBLISHED 09 SEPTEMBER 1975 (SHIMIZU ET AL)	1-6
X	US, A, 4,146,403, PUBLISHED 27 MARCH 1979 (ARMANINI ET AL)	1-6
A	US, A, 4,192,691, PUBLISHED 11 MARCH 1980 (ARMANINI)	1-6
A	US, A, 4,220,675, PUBLISHED 02 SEPTEMBER 1980 (IMAZAKI)	1-6
O	US, A, "USE OF NON-METALLIC PEARLESCENT PIGMENTS TO ACHIEVE METALLIC APPEARANCE", PRESENTED AT 37TH SPE ANNUAL TECHNICAL CONFERENCES, NEW ORLEANS, LA., 09 MAY 1979, RIEGER	1-6
X	US, A, "MICACEOUS IRON OXIDE PIGMENTS", J. OIL COL. CHEM. ASSOC., 1981, 64, pp. 57-74, BISHOP	
<p>* Special categories of cited documents: <sup>15</sup></p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&amp;" document member of the same patent family</p>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search <sup>2</sup>	Date of Mailing of this International Search Report <sup>2</sup>	
08 DECEMBER 1983	19 DEC. 1983	
International Searching Authority <sup>1</sup>	Signature of Authorized Officer	
ISA/US	NATHAN M. NUTTER	

# INTERNATIONAL SEARCH REPORT

International Application No **PCT/US83/01634**

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (if several classification symbols apply, indicate all) <sup>3</sup> According to International Patent Classification (IPC) or to both National Classification and IPC <b>388.3, 388.4, 524/31, 32, 33, 37, 38, 39, 40, 431, 106/288R, 288B, 291, 304, DIG 3</b>		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>4</sup>		
Classification System	Classification Symbols	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>5</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT</b> <sup>14</sup>		
Category <sup>*</sup>	Citation of Document, <sup>16</sup> with indication, where appropriate, of the relevant passages <sup>17</sup>	Relevant to Claim No. <sup>18</sup>
<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p><sup>*</sup> Special categories of cited documents: <sup>15</sup></p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&amp;" document member of the same patent family</p> </div> </div>		
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Date of the Actual Completion of the International Search <sup>1</sup>	Date of Mailing of this International Search Report <sup>1</sup>	
	<div style="font-size: 1.5em; font-weight: bold;">19 DEC. 1983</div>	
International Searching Authority <sup>1</sup>	Signature of Authorized Officer <sup>20</sup>	